$d+is$ vs $d+id'$ Time Reversal Symmetry Breaking States in Finite Size Systems

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We report self-consistent quasiclassical calculations of spontaneous currents and magnetic moments in finite size unconventional superconducting systems, namely: (i) in isolated d-wave superconductor islands where, in addition to the dominant order parameter (with a $d_{x^2-y^2}$ symmetry), a subdominant order parameter of s or d_{xy} symmetry is added; (ii) in grain boundary junctions between two arbitrarily oriented d-wave superconductors, and between a d-wave and an s-wave superconductor. We show that the profile of the spontaneous current density and the magnetic field distribution depend on the time-reversal symmetry breaking properties of the system. For the $d_{x^2-y^2}+id_{xy}$ state, vortices appear near the edges of the finite size systems. We associate these vortices with the chiral nature of the mixed order parameter. The method developed here is quite general, and can be used for predicting properties of any finite size superconducting system.

The possibility of the existence of mixed order parameter symmetries in unconventional superconductors has been suggested and recently investigated both theoretically [1–8] and experimentally [9–14]. A significant feature of the mixed symmetry states is that they may produce spontaneous currents and magnetic moments which can be measured using appropriate experimental techniques. High T_c cuprates present an especially interesting class of materials for studying mixed symmetry states. In these systems, recently developed technology [15] provides an opportunity to fabricate different structures with controllable characteristics, such as π-junctions [1,16] (junctions with equilibrium phase difference equal to π), submicron size ϕ_0-junctions [17] (with an equilibrium phase difference ϕ_0 which is different from 0 or π [5]), π-SQUIDs [1,18] and $\pi/2$-SQUIDs [19], and superconducting qubit prototypes [20]. Therefore, a quantitative prediction of properties of such finite size unconventional superconducting systems becomes extremely important for both experiments and technology.

The mixed symmetry states can be realized near a surface of a d-wave superconductor or at a grain boundary junction between two different superconductors. In the grain boundary junction case, the symmetry of the mixed state is dictated by the symmetries of the order parameters on both sides of the junction (proximity effect) [1–5]. On the other hand, at a surface, a subdominant order parameter may appear due to an attractive interaction potential in the corresponding channel. This may involve a second order phase transition below the superconducting transition temperature [5]. The mixed symmetry states that have been suggested for d-wave superconductors are $d_{x^2-y^2}+id_{xy}$ and $d_{x^2-y^2}+is$ [6] (we denote them $d+id'$ and $d+is$, respectively). Which of these states is realized near the surface of a d-wave superconductor is still an open question. An identification of this state would provide useful information about microscopic interactions in the superconductor. The $d+id'$ and $d+is$ states are inherently different in the sense that the Cooper pairs in the former case have intrinsic magnetic moment, while in the latter, they do not [21,5]. Therefore, it is natural to expect differences in some measurable quantities, such as spontaneous current and magnetic field.

In this article, we perform self-consistent calculations of spontaneous current densities and magnetic field distributions for finite size systems with different mixed symmetries. We show that these experimentally observable characteristics are very dependent on the time-reversal symmetry properties of the system. For the $d+id'$ state, we find that vortices appear, and we connect their appearance with the chiral nature of the order parameter in this state.

Most general approaches to calculation of currents in superconducting systems are based on Gorkov equations for Green's functions of the superconductor. It is widely accepted that this "mean field" approach is valid on a phenomenological level [22], independent of further developments in the first principles' theory of high T_c superconductivity. In a quasiclassical limit, these equations (which are also called Eilenberger equations, see Ref. [23]) can be solved by integrating along the classical trajectories of the quasiparticles (with boundary conditions at infinity). In finite size systems, where the trajectories undergo multiple reflections from surfaces and interfaces, the problem of definition of the boundary conditions becomes complicated. We demonstrate how a modification of the Eilenberger formalism based on the Schopohl-Maki transformation [24] can be used for stable numerical calculations in finite size two-dimensional (2D) systems.

The Eilenberger equations for quasiclassical Green's functions can be written as

$$\mathbf{v}_F \cdot \nabla \widehat{g} + [\omega\widehat{\tau}_3 + \widehat{\Delta}, \widehat{g}] = \widehat{0}, \qquad \widehat{g}^{\,2} = \widehat{1}, \qquad (1)$$

where ω is the Matsubara frequency and

$$\widehat{\tau}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \widehat{g} = \begin{pmatrix} g & f \\ f^\dagger & -g \end{pmatrix}, \quad \widehat{\Delta} = \begin{pmatrix} 0 & \Delta \\ \Delta^\dagger & 0 \end{pmatrix}.$$

The matrix Green's function $\widehat{g}$ and the superconducting order parameter Δ are both functions of the Fermi velocity $\mathbf{v}_F$ and the position $\mathbf{r}$. Δ is determined by the self-consistency equation, which in the two-dimensional (2D) case can be written as

$$\Delta(\theta) = 2\pi N(0)T \sum_{\omega>0} \langle V_{\theta\theta'} f(\theta') \rangle_{\theta'} \qquad (2)$$

where θ is the angle between $\mathbf{v}_F$ and the x-axis, $V_{\theta\theta'}$ is the interaction potential, $N(0)$ is the density of states at the Fermi surface, and $\langle ... \rangle_\theta$ represents averaging over θ. We also have $\Delta^\dagger = \Delta^*$, which holds for any singlet order parameter symmetry. Generally, it is possible to obtain an order parameter which is a mixture of several terms with different symmetries, e.g., $\Delta = \Delta_{x^2-y^2} + \Delta_{xy} + \Delta_s$, where $\Delta_{x^2-y^2} = \Delta_1 \cos 2\theta$, $\Delta_{xy} = \Delta_2 \sin 2\theta$, and Δ_s are the dominant $d_{x^2-y^2}$ component, the subdominant d_{xy}, and the s components of the order parameter, respectively. The corresponding interaction potential,

$$V_{\theta\theta'} = V_{d1}\cos 2\theta \cos 2\theta' + V_{d2}\sin 2\theta \sin 2\theta' + V_s, \quad (3)$$

should be substituted in the self-consistency equation (2) in order to assure a self-consistency of the solution [5]. The current density $\mathbf{j}(\mathbf{r})$ is given by

$$\mathbf{j} = -4\pi i e N(0)T \sum_{\omega>0} \langle \mathbf{v}_F g \rangle_\theta. \qquad (4)$$

For numerical calculations, it is convenient to parameterize the quasiclassical Green's functions via [24]

$$g = \frac{1-ab}{1+ab}, \quad f = \frac{2a}{1+ab}, \quad f^\dagger = \frac{2b}{1+ab}. \qquad (5)$$

Functions a and b satisfy two independent, but nonlinear, (Riccati) equations:

$$\mathbf{v}_F \cdot \nabla a = \Delta - \Delta^* a^2 - 2\omega a,$$
$$-\mathbf{v}_F \cdot \nabla b = \Delta^* - \Delta b^2 - 2\omega b. \qquad (6)$$

From these equations it follows that $a(-\mathbf{v}_F) = b^*(\mathbf{v}_F)$ and $b(-\mathbf{v}_F) = a^*(\mathbf{v}_F)$. The solutions of these equations can be interpreted as *trajectories* of quasiparticles. One should integrate these equations along all possible trajectories and perform the summation over the trajectories to calculate the current. To find a and b along the trajectories, one needs to use boundary conditions at the ends of the trajectories. In infinite systems, one usually assumes that *all* trajectories go deep into the bulk of the superconductor, i.e., one can use the bulk solutions

$$a_\pm = \frac{\Delta}{\omega \pm \Omega}, \qquad b_\pm = \frac{\Delta^*}{\omega \pm \Omega}, \qquad (7)$$

with $\Omega = \sqrt{\omega^2 + |\Delta|^2}$, as the boundary conditions at infinity. In the case of finite size systems where multiple reflections from surfaces and interfaces are possible, stability of the numerical procedure for calculating the current is not obvious because of complications with choosing a proper boundary condition for a given trajectory. Nevertheless, we show that a stable numerical procedure can still be developed.

Numerical integration for a (b) in Eq. (6) should be taken in the direction of $\mathbf{v}_F$ ($-\mathbf{v}_F$) to ensure stability. When Δ is a constant, the solution for a can be written analytically,

$$a_f = a_+ + \frac{a_i - a_+}{1 + \frac{\Delta^*}{\Omega}(a_i - a_-)e^{\Omega\tau}\sinh\Omega\tau} \qquad (8)$$

$$\approx a_+ + \frac{\Omega}{\Delta^*}\left(\frac{a_i - a_+}{a_i - a_-}\right)e^{-2\Omega\tau} \qquad \text{(if } \Omega\tau \gg 1),$$

where a_i and a_f are the values of a at the initial ($\mathbf{r}_i$) and final ($\mathbf{r}_f$) points of the trajectory, and $\tau = |\mathbf{r}_f - \mathbf{r}_i|/v_F$ is the migration time between the initial and final points. It is clear that the solution for a relaxes to the bulk value a_+ at the distance $L = \mathbf{v}_F/2\Omega$ which is of the order of the coherence length ξ_0. In other words, when the quasiparticle moves away from the initial point at a distance of a few ξ_0's, any information about the initial point a_i is lost. The same argument is valid for the function b.

Let us now consider a system with a restricted geometry. After integrating over a few ξ_0's (considering the reflections), a_f becomes almost independent of a_i. This solution corresponds to a simple exponential relaxation of the functions a and b to their local "steady-state" values (which can be different from the bulk values) defined by the local values of the order parameter. Therefore, to find Δ at a given point, one does not need the values of Δ at distances larger than several ξ_0 along the trajectory. Such a "relaxational" property of the a and b functions significantly simplifies the numerical solution of the self-consistent 2D problem. All the "multiple-reflection history" of a trajectory becomes a moot point, and for practical calculations, this trajectory can be cut at a distance of a few ξ_0's from a given point. We used this observation in our self-consistent calculations. For the "cutting" distance, we choose $10\xi_0$–$20\xi_0$. We set the bulk values of a and b (a_+ and b_+) calculated at the initial point of the "truncated" trajectory as the boundary conditions (such a choice does not affect the final results because the system has no memory) and integrate along the trajectory until we get to the point where we calculate the current. We found that the results are very stable and do not depend on the value of the "cutting" distance.

To compare the spontaneous currents and magnetic fields in the $d + is$ and $d + id'$ states, we performed self-consistent calculations of the order parameter in small size triangular regions of clean d-wave superconductor with different subdominant order parameters. We assume specular boundaries with the length of the longer edge of the triangles being $30\xi_0$. Figs. 1 and 2 show the spontaneous current and magnetic field distributions
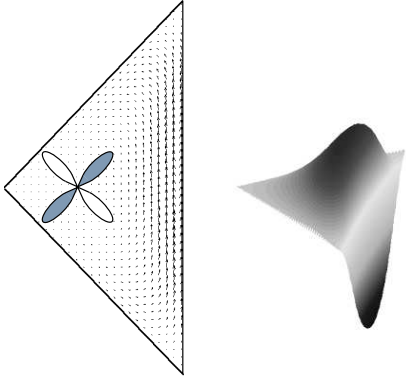
FIG. 1. Spontaneous current (left) and magnetic field (right) in a finite size triangular-shaped region of a d-wave superconductor with a subdominant s-wave, at the temperature $T = 0.2T_{cs} = 0.1T_c$. The orientation of the dominant $d_{x^2-y^2}$ order parameter is shown.
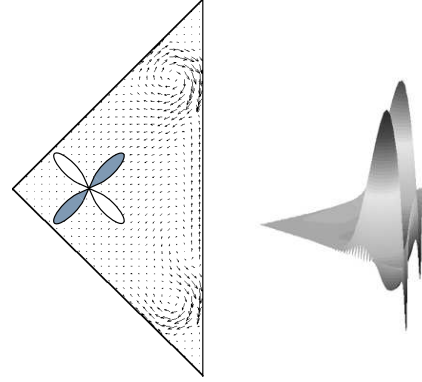

FIG. 2. The results of self-consistent calculations similar to those of Fig. 1 but with a subdominant d_{xy}-wave order parameter, and at the temperature $T = 0.2T_{c2} = 0.1T_c$.

in systems with the two different symmetries. We imposed the desired subdominant symmetry by introducing an additional attractive interaction potential in the corresponding channel [V_{d2} or V_s in Eq. (3)]. The subdominant critical temperatures are taken to be $0.5T_c$ in both cases. The orientation of the dominant order parameter makes a 45° angle with respect to the longer (right) edge of the triangle (see the figures). As a result, only this boundary of the triangle is pair breaking; the quasiparticles face a different sign of the dominant order parameter after reflection from the surface [5]. The suppression of the main order parameter at the surface allows the subdominant order parameter to appear near the surface.

The spontaneous current and magnetic field distributions are evidently different in Figs. 1 and 2. In the first case, the current makes a counterclockwise loop throughout the region, while in the second case, the direction of rotation of the current changes throughout the triangle. Especially, appearance of the two vortices in Fig. 2 reflects the chiral nature of the $d + id'$ symmetry of the order parameter [5]. There is no phase winding around these vortices and therefore no flux quantization is expected. In fact, the flux trapped in these vortices is much smaller than a flux quantum. The magnitude of the magnetic field at the maximum positions is of the order of 10^{-4}–$10^{-3} G$ in both cases. This value agrees with the magnitude of the magnetic field reported in Ref. [12]. In the $d + id'$ case, the magnetic field is strongly peaked at the vortices. The magnetic field in other spatial points is noticeable only in the vicinity of the pair-breaking edge (within a few coherence lengths) and is almost one order of magnitude weaker than the field at the vortex peaks. This is a result of the superscreening effect which happens when the spontaneous current is due to the intrinsic angular moment of the order parameter [4,5]. The total flux generated by this magnetic field is very small and

difficult to measure. This is in agreement with the magnitude of the flux, calculated using a different technique, in Ref. [25]. In the $d + is$ case, on the other hand, the peak is spread over a wider region. In fact, in larger systems, the size of this region could be of the order of the penetration depth. This is because in the $d + is$ case, the superscreening effect is absent and the only mechanism to return the current is the Meissner effect, which happens over the length scale of a penetration depth. The flux generated by this magnetic field can be large and, therefore, measurable. This suggests that the flux measured in Ref. [12] can be generated by a $d + is$ symmetry breaking state and not by a $d + id'$ one.

The spontaneous magnetic moments of mesoscopic islands with $d + is$ or $d + id'$ symmetry can be in principle used as qubits. Since the "up" and "down" states are degenerate eigenstates of the effective Hamiltonian, tunneling between them becomes only possible in the presence of external magnetic field, field gradient, etc., depending on the multipole structure of the spontaneous current distribution. In order to determine the tunneling amplitude, a three-dimensional picture should be considered, which is beyond the scope of this publication.

We also have calculated the spontaneous current and magnetic field distributions in grain boundary junctions. The geometry of these systems consists of a square of size $= 30\xi_0 \times 30\xi_0$, divided into two equal parts separated by a grain boundary junction. The right half of the square is a clean d-wave superconductor with a 45° orientation of the order parameter with respect to the boundaries. The left half, on the other hand, can be either a clean s-wave superconductor (s-d junction, Fig. 3), or a clean d-wave superconductor with a 0°-orientation (d-d junction, Fig. 4). The grain boundary is taken to be perfectly transparent with no roughness or faceting [26]. Although this choice of grain boundary does not exactly correspond to the reality, study of it may provide useful information for real systems. To be able to compare the two systems, we assumed that the s-wave and d-wave

superconductors have the same transition temperatures. We did not introduce any subdominant order parameter here. However, we introduced an additional phase difference of $\Delta\phi = \pi/2$ between the two sides. Such a choice corresponds to the equilibrium phase difference of the junction at which the total current passing through the junction is zero [5]. Calculations are done at $T = 0.1T_c$.

The results of spontaneous current density and magnetic field distributions are shown in Fig. 3 for the s-d junction, and Fig. 4 for the d-d junction. Note that the current distribution is not symmetric with respect to the grain boundary. For the s-d junction of Fig. 3, the current has a maximum at the grain boundary and returns through the bulk of the superconductors on both sides. If the system is large, the size of the region where the spontaneous current is non-zero, should be of order of the penetration depth. For the d-d junction (Fig. 4), on the other hand, the current changes the direction just within a few coherence length from the boundary, again due to the superscreening effect [4,5]. Notice also that near the edges of the system, on the left side of the grain boundary junction ($0°$-orientation of the order parameter), the current returns along the diagonal, whereas on the right side ($45°$-orientation) it forms two small vortices and antivortices. These vortices, which are absent in the s-d case, again reflect the chiral nature of the $d + id'$ symmetry; although the subdominant order parameter is absent in this case, the correlation functions convey this symmetry near the grain boundary junction due to the proximity effect [5]. The magnetic field distributions, displayed in Figs. 3 and 4, is of the order of $10^{-4}G$ at the positions of the maxima, in both s-d and d-d junctions. In the case of d-d junction (Fig. 4), magnetic field is peaked at the location of vortices. Away from the vortices, the magnetic field is localized near the grain boundary junction and almost one order of magnitude smaller. The flux generated by such a magnetic field is very small. Thus, observation of large flux as in Ref. [10] can not be associated with such an effect. In real systems, due to the faceting effect [26], some π-loops may exist along the grain boundary junction. They can produce large fluxes, of the same order as observed in Ref. [10].

It is important to emphasize here that the existence of the spontaneous current at the grain boundary junctions does not depend on the presence of a subdominant order parameter (unlike in the previous case, unless if we consider other effects [27]). Thus, experiments similar to that of Ref. [11] (that only probes the mixing of the symmetry of the order parameter) can not exclude the possibility of the existence of spontaneous current. Indeed, addition of a subdominant order parameter will suppress the spontaneous current at the boundary (see Refs. [4,5]).

One should note that in the presence of magnetic field, the procedure described here is not valid, because a path dependent phase will be accumulated to a and b functions, and the relaxation mechanism along the trajectory may no longer hold. This does not mean that we cannot
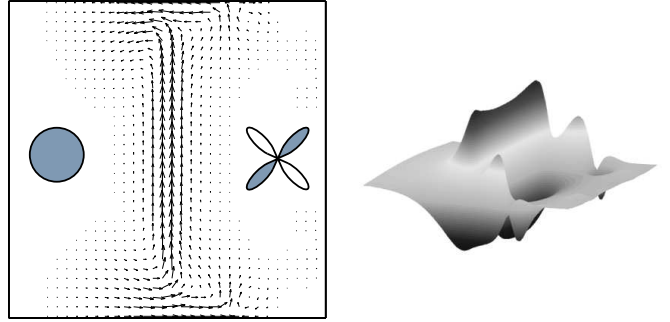

FIG. 3. Spontaneous current density (left) and magnetic field (right) distributions for an s-d junction. The grain boundary is a vertical line located in the middle.
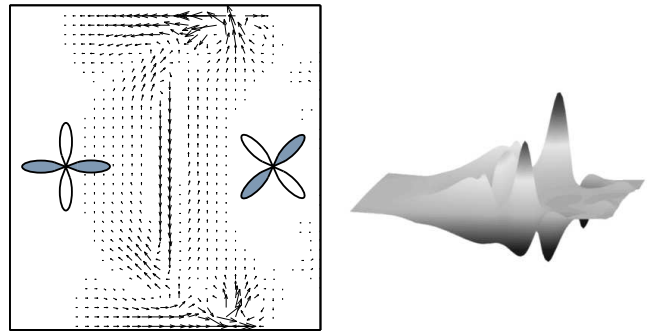

FIG. 4. Spontaneous current density (left) and magnetic field (right) distributions for a grain boundary junction between two d-wave superconductors.

use our approach to calculate currents and fields in realistic systems without calculating the magnetic field due to spontaneous currents self-consistently. Indeed, it is not difficult to show that the corresponding Doppler shift is rather small. We saw that for systems with size $L \sim 30\xi_0 \sim 5 \times 10^4$Å, spontaneous magnetic field is always smaller than 10^{-3}G. Therefore, the induced superfluid momentum is at most $p_s \sim (e/c)HL \sim 10^{-26}$g $\cdot$ cm/s. If we take $\hbar v_F \simeq 1$ eV$\cdot$Å [28] for the Fermi velocity in high-T_c cuprates, we obtain $p_s v_F \sim 0.1$ μeV ~ 1 mK, which is negligible compared to other energy scales in the problem [29]. However, these restrictions may be important if the system is placed in a strong external magnetic field.

To summarize, we described self-consistent calculations of some equilibrium properties in finite size superconducting systems. We calculated distributions of spontaneous current and magnetic field in different small samples in the presence of mixed order parameter symmetries. The nature of the mixed symmetry state in all cases affects the shape of the current and magnetic field distributions. In particular, the chiral nature of the $d+id'$ states exhibits itself through the appearance of vortices close to the edges of the system. The vortices are absent in the $d + is$ cases. The method described here is

quite general and can be applied to any 2D geometry with proper boundary conditions as long as external magnetic field is not present. The self-generated magnetic field by the spontaneous currents, however, is usually very small, i.e., it can be neglected in most of the practically important cases.

[1] M. Sigrist and T.M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992); M. Sigrist, D.B. Bailey, and R.B. Laughlin, Phys. Rev. Lett. **74**, 3249 (1995); M. Sigrist, Prog. Theor. Phys. **99**, 899 (1998).

[2] A. Huck, A. van Otterlo, and M. Sigrist, Phys. Rev. B **56**, 14163 (1997).

[3] Yu.S. Barash, A.V. Galaktionov, and A.D. Zaikin, Phys. Rev. B **52**, 665 (1995).

[4] M.H.S. Amin, A.N. Omelyanchouk, and A.M. Zagoskin, Phys. Rev. B **63**, 212502 (2001).

[5] M.H.S. Amin, A.N. Omelyanchouk, S.N. Rashkeev, M. Coury, and A.M. Zagoskin, Physica B **318**, 162 (2002).

[6] M. Matsumoto and H. Shiba, J. Phys. Soc. Japan **64**, 1703 (1995); **64**, 3384 (1995); **64**, 4867 (1995); **65** 2194 (1996).

[7] S. Yip, Physical Review B **52**, 3087 (1995).

[8] M. Fogelström, D. Rainer, and J.A. Sauls, Phys. Rev. Lett. **79**, 281 (1997); M. Fogelström and S.-K. Yip, Phys. Rev. B **57**, R14060 (1998).

[9] M. Covington, M. Aprili, E. Paraoanu, L.H. Greene, F. Xu, J. Zhu, and C.A. Mirkin, Phys. Rev. Lett. **79**, 277 (1997); L.H. Greene, M. Covington, M. Aprili, E. Badica, and D.E. Pugel, Physica B **280**, 159 (2000).

[10] J. Mannhart, *et al.* Phys. Rev. Lett. **77**, 2782 (1996).

[11] W.K. Neils and D.J. Van Harlingen, Phys. Rev. Lett. **88**, 047001 (2002).

[12] R. Carmi, E. Polturak, G. Koren, and A. Auerbach, Nature **396**, 168 (1998).

[13] F. Tafuri and J.R. Kirtley, Phys. Rev. B **62**, 13934 (2000).

[14] H. Walter *et al.* Phys. Rev. Lett. **80**, 3598 (1998).

[15] A.Yu. Tzalenchuk, *et al.*, preprint.

[16] V.B. Geshkenbein, A.I. Larkin, and A. Barone, Phys. Rev. B **36**, 235 (1987); L.N. Bulaevskii, V.V. Kuzii, and A.A. Sobyanin, JETP Lett. **25** 290 (1977).

[17] E. Iĺichev, *et al.* Phys. Rev. Lett. **86**, 5369 (2001).

[18] R.R. Schulz, *et al.*, Appl. Phys. Lett. **76**, 912 (2000).

[19] M.H.S. Amin, M. Coury, and G. Rose, IEEE Trans. Appl. Supercond., in press (cond-mat/0107370).

[20] L.B. Ioffe, *et al.*, Nature **398**, 679 (1999); A. Blais and A.M. Zagoskin, Phys. Rev. A **61**, 042308 (2000).

[21] G. E. Volovik, JETP Lett., **66**, 522 (1997).

[22] V.P. Mineev and K.V. Samokhin, *Introduction to unconventional superconductivity*, Gordon and Breach Science Publishers (1999).

[23] G. Eilenberger, Z. Phys. **214**, 195 (1968).

[24] N. Schopohl and K. Maki, Phys. Rev. B **52**, 490 (1995); N. Schopohl, preprint (cond-mat/9804064).

[25] J.-X. Zhu and C.S. Ting, Phys. Rev. B **60**, R3739 (1999).

[26] H. Hilgenkamp, J. Mannhart, and B. Mayer, Phys. Rev. B **53**, 14586 (1996).

[27] T. Löfwander, V.S. Shumeiko, and G. Wendin, Phys. Rev. B **62**, R14653 (2000).

[28] A.V. Balatsky and P. Bourges, Phys. Rev. Lett. **82**, 5337 (1999) (see note to Ref.[28]).

[29] M. Aprili, E. Badica, and L.H. Greene, Phys. Rev. Lett. **83**, 4630 (1999).